

Supporting Information

Continuous Hydrothermal Flow Synthesis of Graphene Quantum Dots

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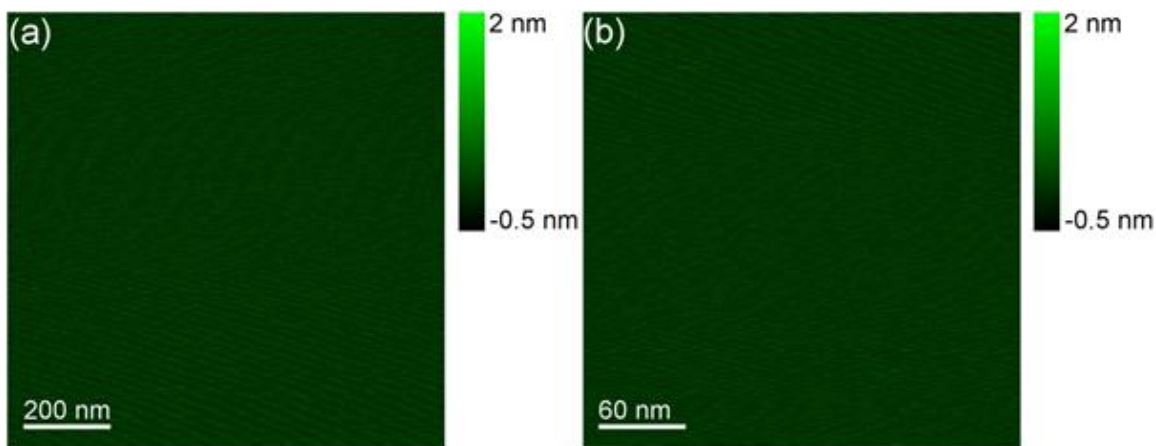


Figure S1: AFM height images at scan sizes of 1 x 1 μm (a) and 300 x 300 nm (b) of freshly cleaved mica showing an atomically flat and clean surface prior to GQD spin coating deposition.

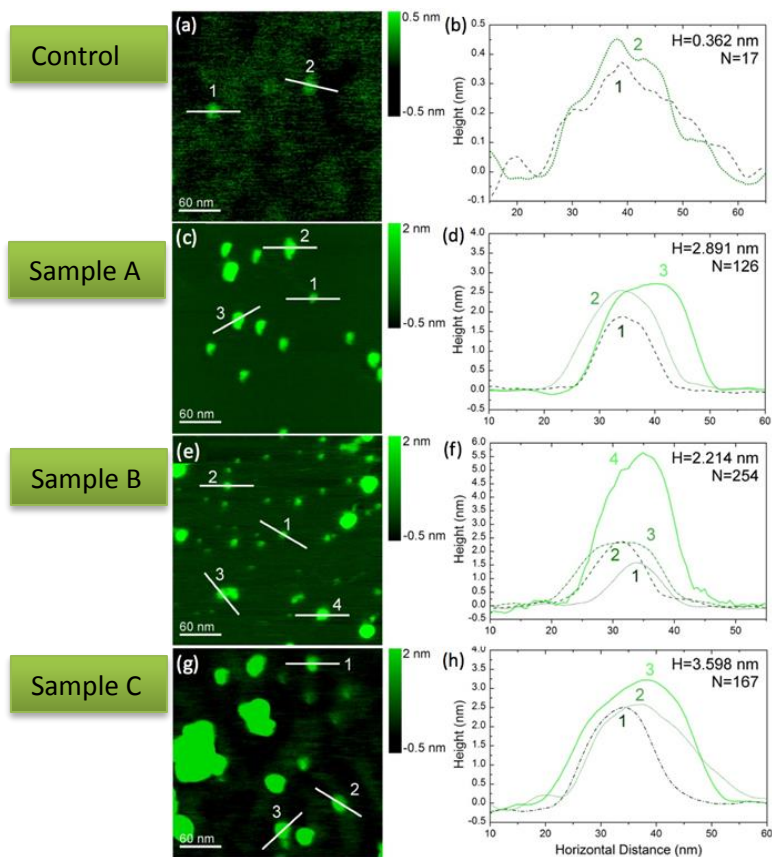


Figure S2: AFM height images at scan sizes of 1 x 1 μm and 300 x 300 nm, and corresponding cross sections for GQDs (“Control”) (a) – (b), and GQDs with varying amounts of PCX4 (“Samples A – C”) (c)-(h), respectively. Average heights (H) and sample numbers (N) are given in the top right corner of each cross-section graph.

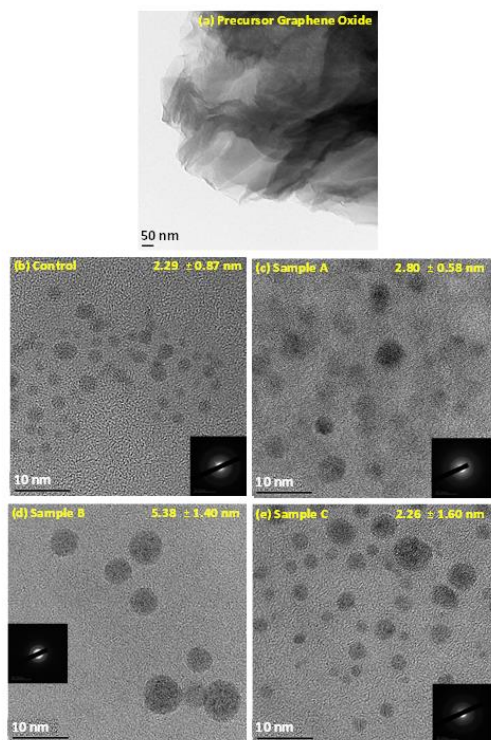
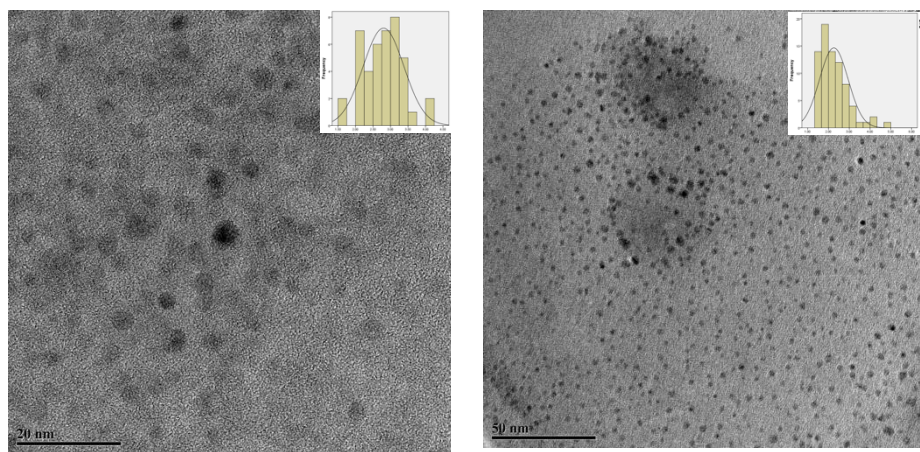


Figure S3: TEM images of (a) precursor graphene oxide (b) Control, (c) Sample A, (d) Sample B, (e) Sample C.



Additional TEM images (left) Sample A (right) sample C.

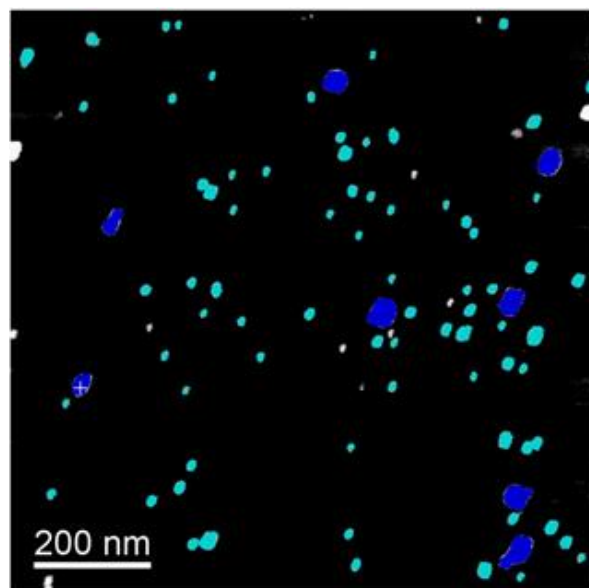


Figure S4: Particle analysis was conducted using the NanoScope Analysis software (version 1.4). The software identified all particles on the surface and then clusters were manually removed from the sample group in order to accurately determine the height of individualised nanoparticles. An example of particle selection is seen above, where only individualised GDQs (light blue) were measured and clusters (dark blue), which were identified with the aid of peak force error images, were excluded. An example of Peak force images for cluster identification is seen in Figure S5.

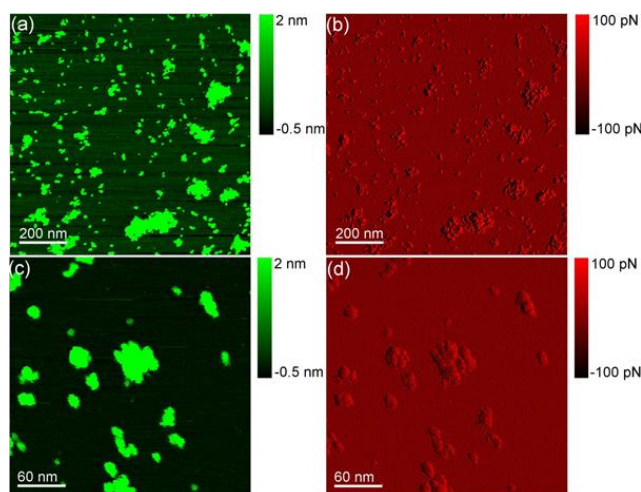


Figure S5: AFM height ((a) and (c)) and corresponding peak force error ((b) and (d)) images at scan sizes of 1 x 1 μm and (b) 300 x 300 nm, respectively. Peak force error measures the deviation from the peak force set point (500 pN) as a result of slow reaction by the system when topography is traversed. As most deviation occurs at areas of dramatically changing topography, it accentuates edges and can be used to identify clusters, which consist of multiple GDQs.

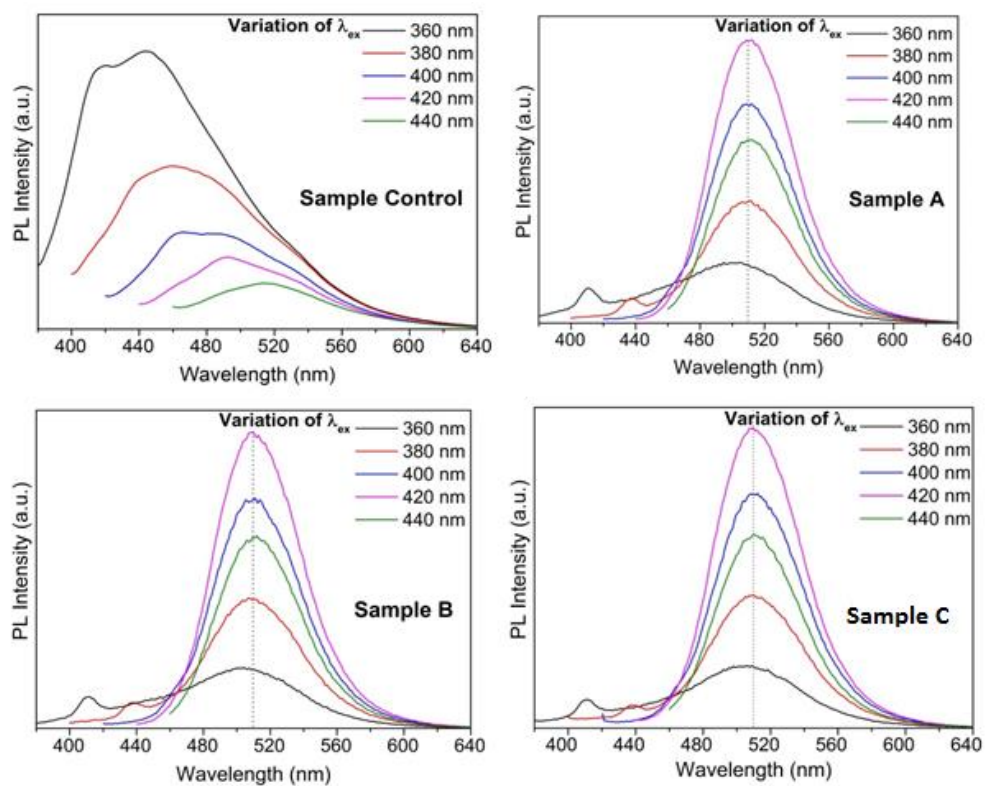


Figure S6: PL emission spectra of GQD recorded at different excitation wavelength (360-440 nm, 20 nm intervals).

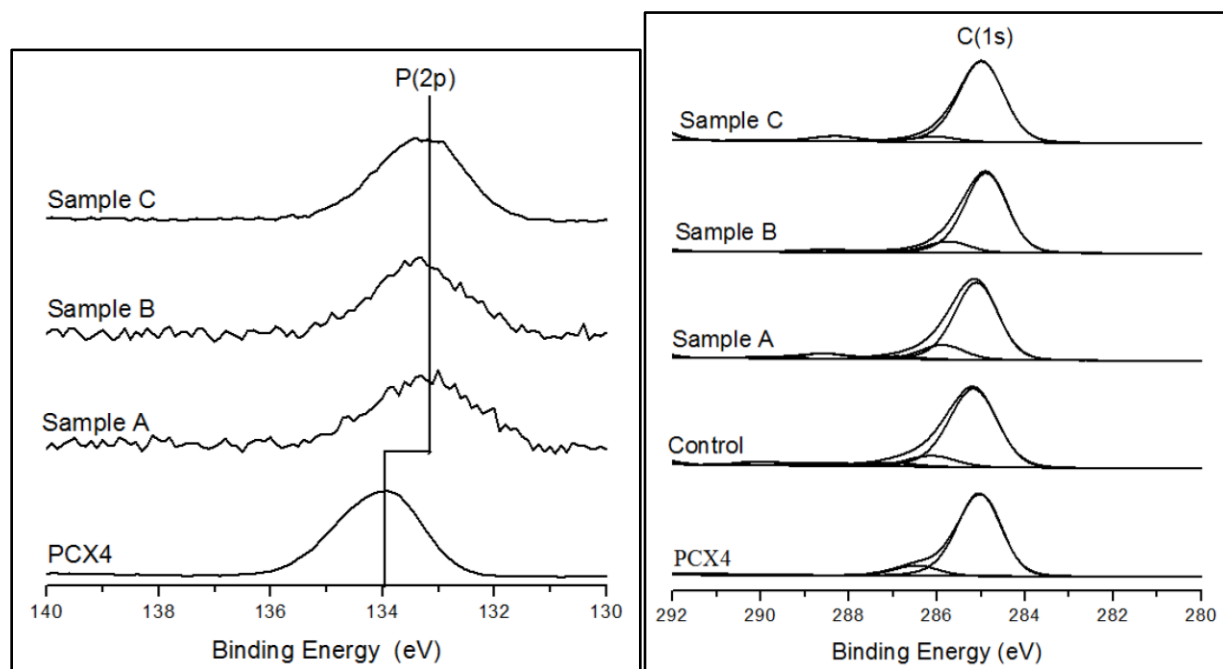


Figure S7: XPS Spectra

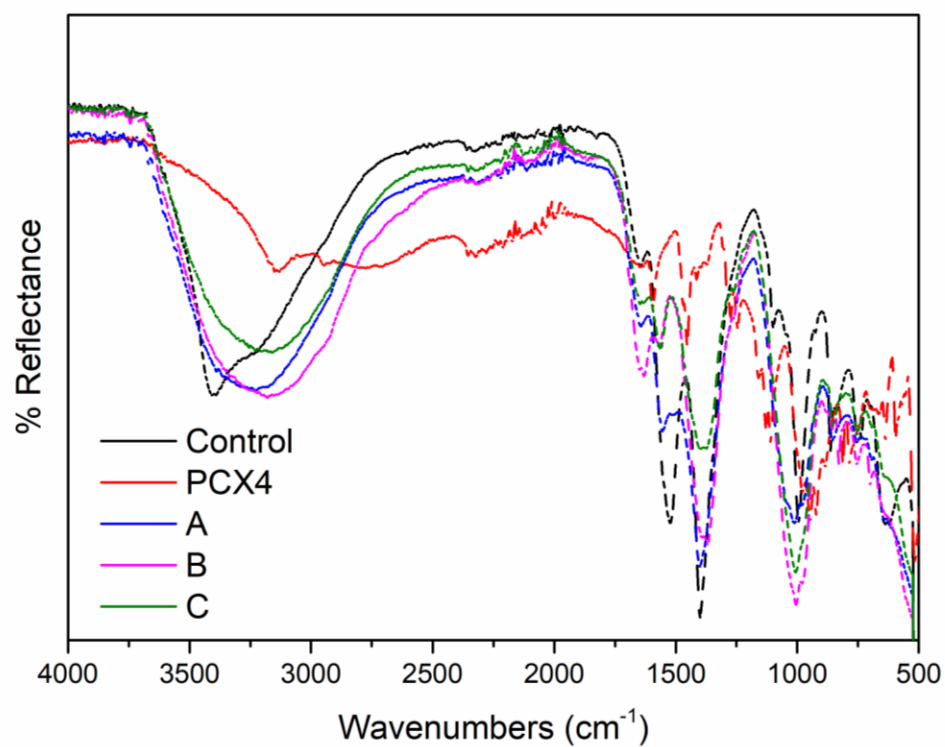


Figure S8: FT-IR Spectra.

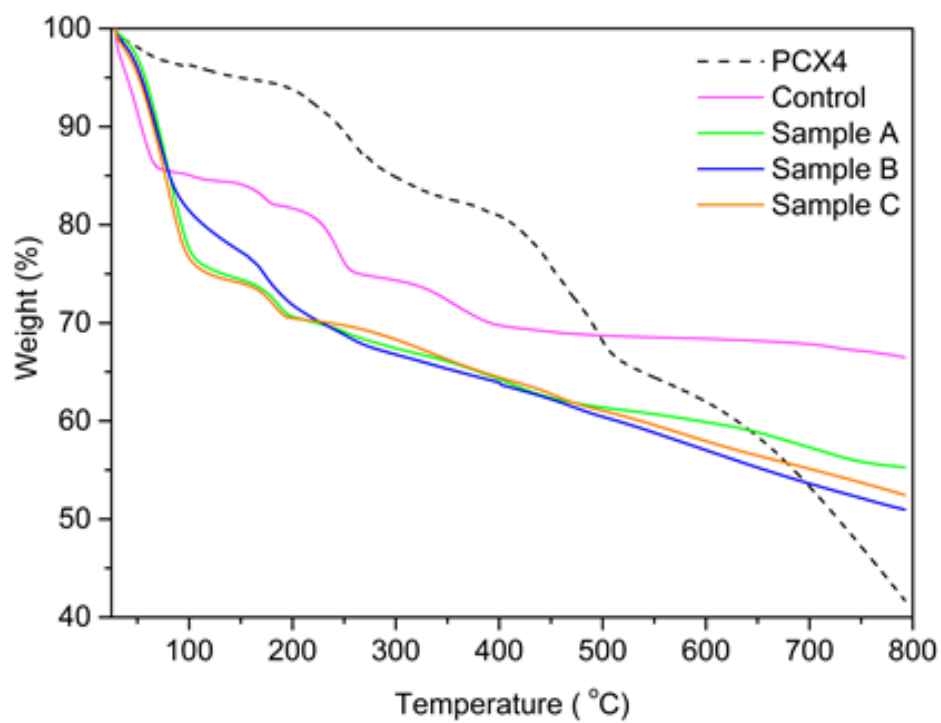


Figure S9: TGA diagrams.

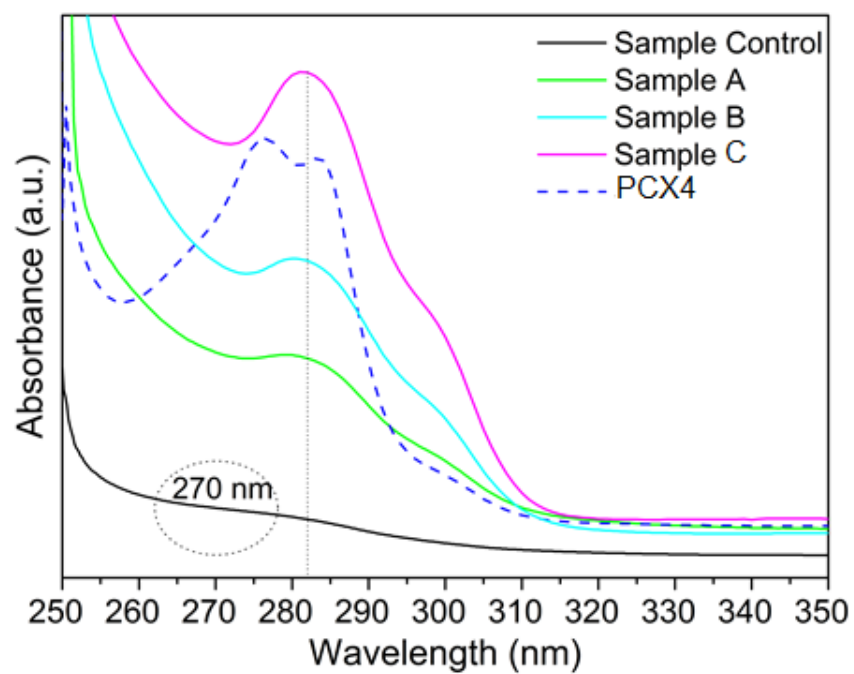
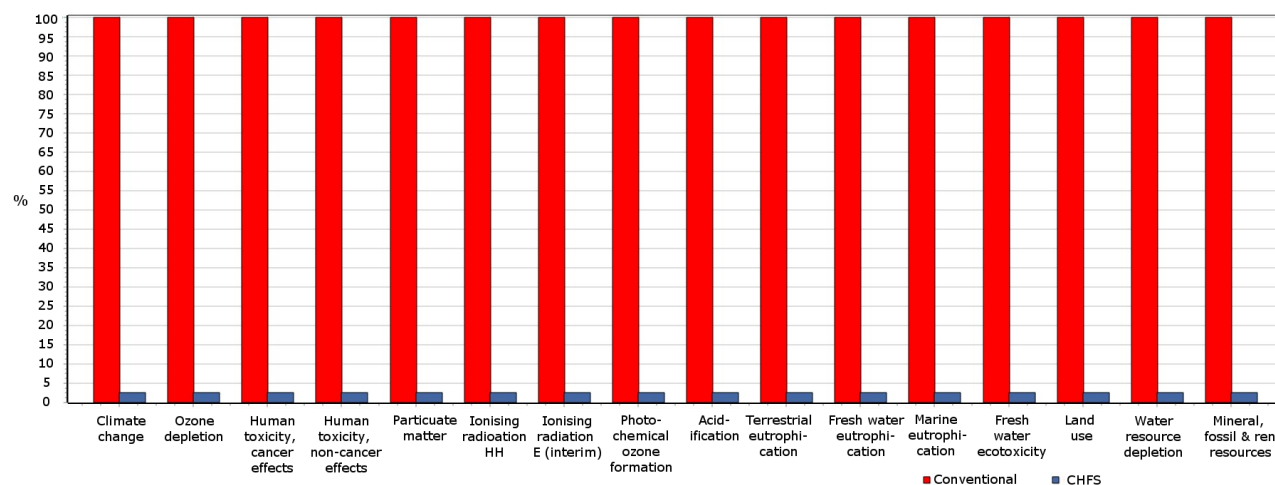


Figure S10: UV-Vis Spectra.

Table S1: Life cycle inventories to produce 1 mg GQDs including the estimated energy consumption of the laboratory equipment. Legend key: Red – conventional hydrothermal batch synthesis; Blue- Continuous Hydrothermal Flow Synthesis (CHFS).



Conventional batch syntheses				
Materials/fuels				
Nitric acid, without water, in 50% solution state (GLO) market for Alloc Rec, U	2.62	g	2.6 M nitric acid, 40 mL, 6.55 g /2.5	
Water, deionised, from tap water, at user (Europe without Switzerland) market for water Alloc Rec, U	16	g	40 g /2.5	
Sodium hydroxide, without water, in 50% solution state (GLO) market for Alloc Rec, U	1.67	g	NaOH 4.16 g /2.5	
Electricity/heat				
Electricity, low voltage (RER) market group for Alloc Rec, U	1.68	kWh	heater 500W, reflux @ 70C, 24h	
Electricity, low voltage (RER) market group for Alloc Rec, U	4.8	kWh	autoclave heater 500W @ 200C, 24h	
Electricity, low voltage (RER) market group for Alloc Rec, U	0.05	kWh	sonificator 100W, 30 min	
Continuous Hydrothermal Flow Syntheses (CHFS)				
Materials/fuels				
Potassium hydroxide (GLO) market for Alloc Rec, U	0.44	g	KOH	
Water, deionised, from tap water, at user (Europe without Switzerland) market for water Alloc Rec, U	240	g		
Electricity/heat				
Electricity, low voltage (RER) market group for Alloc Rec, U	0.03	kWh	3 pumps of 60W, 10 min=0.1667hrs 0,01kWh per pump	
Electricity, low voltage (RER) market group for Alloc Rec, U	0.125	kWh	heater 1000W(750W), 10 min@450°C	
Electricity, low voltage (RER) market group for Alloc Rec, U	0.05	kWh	sonificator 100W, 30 min	

Figure S11: Energy consumption of the laboratory equipment for both the conventional (batch) and CHFS processes according to Table S1.

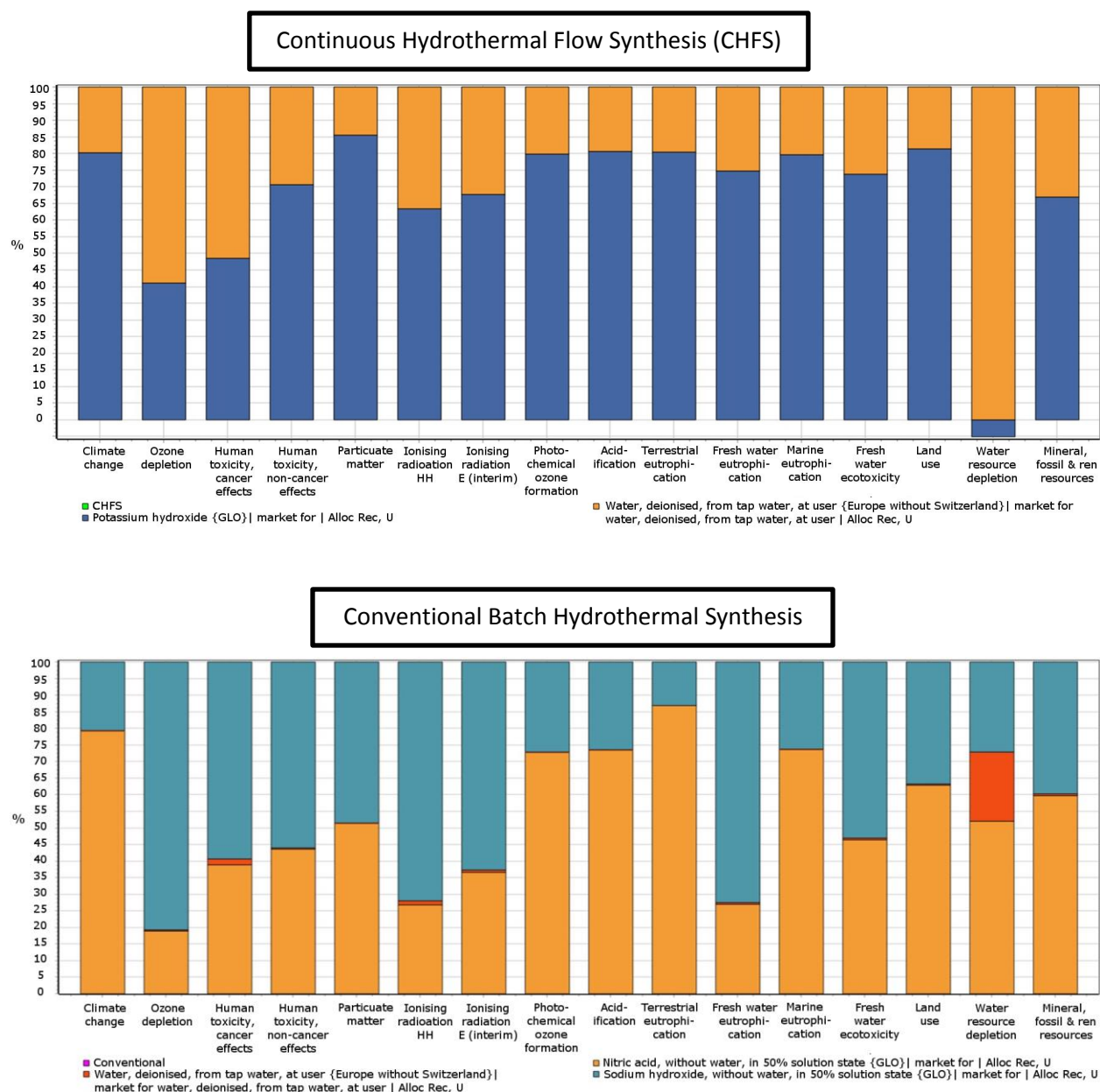


Figure S12: Environmental impact results for two synthesis routes (CHFS versus batch synthesis). For each synthesis type all impact scores have been scaled to 100% (impacts ascribed to the precursors used refer to a proportion of a total impact rather than a quantity). Water resources depletion having the greatest environmental impact for CHFS. Other than that, KOH dominates all other environmental impact categories. For the batch process, the use of sodium hydroxide and nitric acid accounted for a larger proportion of the life cycle impact contribution in most of the categories, varying between 15 - 85% of the impact.